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ScienceDirect

Energy Procedia 4 (2011) 1168–1175

Energy Procedia

www.elsevier.com/locate/procedia

GHGT-10

Integrating IT-SOFC and gasification combined cycle with methanation reactor and hydrogen firing for near zero-emission power generation from coal

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Abstract

Application of Solid Oxide Fuel Cells (SOFC) in gasification-based power plants would represent a turning point in the power generation sector, allowing to considerably increase the electric efficiency of coal-fired power stations while reducing CO_2 and other pollutant emissions. The aim of this paper is the thermodynamic assessment of a SOFC-based IGFC plant with methanation reactor, hydrogen post-firing and CO_2 capture by physical absorption. The configuration proposed allows to obtain a very high net efficiency (51.6%), overcoming the main limits of configurations assessed in previous works.

Keywords: IGFC; gasification; fuel cell; SOFC; methanation; absorption; CO_2 capture

1. Introduction

Application of fuel cells in coal fired power plants can be the key for the achievement of very high conversion efficiencies with extremely low pollutants emissions. Important international projects are focusing on the development of advanced power cycles using fuel cells and gas turbines integrated with a coal gasification plant (e.g. the FutureGen Vision21 projects of the US DOE [1,2]). R&D activities are pushed by the exploitation of a low cost fuel and by the perspective of applying such technology (Integrated Gasification Fuel Cell cycle – IGFC) to high efficiency electricity generation with carbon dioxide capture and storage (CCS). A distinctive advantage of this concept is given by the electrochemical oxidation of the syngas occurring in the fuel cell, which acts like an oxygen combustor avoiding the dilution of exhaust gases with nitrogen.

Some examples of studies on IGFC systems have been referenced in [3], to which we address the reader for a review. Starting from the results of previous works, dealing with IGFCs based on high temperature SOFC [3-5], the aim of the present work is the investigation of a cycle layout based on the integration of coal gasification with

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doi:10.1016/j.egypro.2011.01.170

intermediate temperature SOFCs (IT SOFC, operating at 800°C) and a combined cycle, arranged to maximize the net electrical efficiency when including the CO_2 capture process.

The proposed cycle arrangement is based on a simple cycle gas turbine and a steam bottoming cycle to optimize heat recovery and maximize efficiency, trying to maximize also the power output of lower specific cost components (gas and steam turbines) without including components too far from the present state-of-the-art, apart from the SOFC (whose economic applicability in such configurations and size is still to be proven). The paper discusses the results in terms of energy balances and preliminary technical and economical remarks.

2. Thermodynamics of SOFC-based IGFC cycles

Hybrid SOFC-GT cycles proposed in literature in recent years are generally fuelled with natural gas, which is the base fuel for high temperature fuel cells. The main difference occurring when a CO/H_2-based syngas is used as SOFC fuel instead of natural gas is the lower conversion efficiency and the larger air flow rate required for fuel cell cooling. When methane is present in the fuel gas, heat generated in the fuel cell can be partly used for the endothermic internal reforming and hence efficiently recovered as chemical energy, which can be re-converted into electricity within the fuel cell itself. Conversely, when no methane is present in the fuel gas, air is the only possible coolant and sensible heat collected by the air stream can be only recovered in the bottoming gas and steam cycles. This typically yields an efficiency penalty on the FC conversion efficiency [4], whose effects on the overall energy balances depends on the cycle arrangement and the mutual interaction between the plant components, as already investigated in previous works analyzing MCFC hybrid cycles [6].

Integration of the power cycle with the chemical island where syngas is produced from a solid feedstock is another point characterizing IGFC cycles. A good integration for optimal heat recovery from each plant unit is important to obtain high plant efficiencies. On the basis of previous studies carried out by the authors, the following considerations can be made, useful when designing an IGFC plant layout:

- High fuel cell operating pressures favor overall plant efficiency [4,5]. Beneficial effects resulting from high SOFC voltages compensate the higher than optimal gas turbine pressure ratio (for the typical resulting turbine inlet temperatures). For this reason, it is preferable for the fuel cell pressure to be coupled with the gasification island without intermediate syngas expansions. The resulting FC operating pressure is much higher than in usual SOFC-GT hybrid cycles (20-35 bar vs. 4-10 bar), but the development of SOFCs capable of operating at such pressures does not appear a real barrier for mid-long term plants such IGFCs.
- An optimal fuel utilization factor, which maximizes overall plant efficiency, exists for the fuel cell [5,3]. On one side, with high fuel utilization, high fractions of fuel are converted in the SOFC with the high efficiencies typical of hybrid cycles [7]. A higher water content at anode outlet is another consequence of high fuel utilizations, which lead to reduced anode recycle rate or steam addition required for fuel humidification to avoid carbon deposition. On the other side, low fuel utilizations lead to higher hydrogen contents at fuel cell outlet and consequently higher voltages[1]. More important, lower air flow rate are required for fuel cell cooling and greater combustible species result at anode exhaust. Consequently, higher turbine inlet temperatures are obtained after combustion of the fuel cell outlet streams, positively affecting the efficiency of the bottoming cycle.
- Bypassing part of the syngas directly to the GT combustor has positive effects on both plant economics and efficiency [5]. Contribution of fuel cell (the highest cost power generation component) on plant gross power production reduces and gas turbine specific work increases substantially (i.e. more power is generated by the gas turbine with a given air flow rate). Both these factors positively affect plant economics. Also efficiency increases moderately by burning part of the syngas in the GT combustor, bypassing the fuel cell. This apparently surprising effect can be explained by considering the high

[1] All the cases in [3-5] were calculated by assuming to operate at a constant current density, equal to the economic optimal one, and by correcting the FC potential assumed for a reference case according to a Nernst type equation.

temperature of fuel cell exhausts reacting in the GT combustor and the thermodynamic quality of the combined cycle which recovers energy from the cell exhaust stream. In fact, bypass fuel is used to heat the fuel cell exhausts from the SOFC operating temperature (800°C) up to the state-of-the-art Turbine Inlet Temperature (TIT, in the range 1300-1400°C) and heat of combustion is introduced in the bottoming combined cycle at high temperature and hence converted with a higher than usual average efficiency.

In a recent work analyzing IGFC plants with CO_2 capture [3], based on oxycombustion of SOFC anode exhaust gas (Fig.1a), net efficiencies of 47.1-47.5% were obtained, with penalties of about 6% points with respect to an IGFC cycle without CO_2 capture. The largest weakness of that configuration was probably the very low temperature of the cathode exhaust air, expanded in the gas turbine with no further combustion. Any combustion with anode gas or bypassed syngas is in fact not feasible because it would lead to relevant CO_2 emissions. As a result, sensible heat collected in the SOFC by the relevant air flow is converted in the GT cycle with an extremely low efficiency and specific work.

The plant layout proposed in this work is defined to overcome these limits, by introducing two new processes (Fig.1b):

(i) a methanation process, aimed at increasing the methane content in the fuel gas and hence reducing the air flow rate needed for SOFC cooling and improving the energy conversion efficiency of the integrated power cycle; and

(ii) a hydrogen firing before the gas turbine, using a post-SOFC absorption process for CO_2 capture and for the recovery of the hydrogen not oxidized in the fuel cell, which can be used as fuel for the gas turbine cycle, increasing substantially the TIT.

Two water gas shift (WGS) reactors and a pre-SOFC CO_2 absorption process were also included in the plant in order to obtain an admissible H_2/CO ratio at methanation reactor inlet and to convert CO in the anode exhaust stream into CO_2 and H_2.

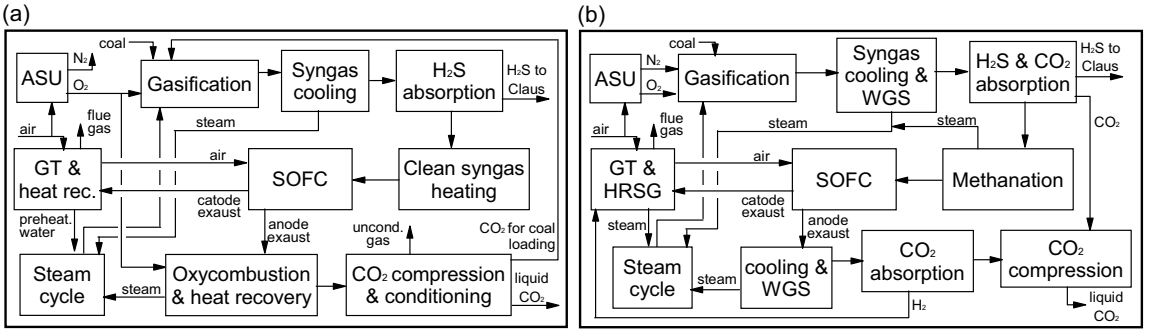

Figure 1: Conceptual layout of proposed IGFC plants with CO_2 capture based on (a) anode exhaust oxycombustion [3] and (b) syngas methanation and hydrogen firing.

3. Plant layout and assumptions

The detailed layout of the assessed plant is shown in Fig.2. Gasification is based on a dry-feed, oxygen-blown, entrained flow Shell type gasifier operating at 44 bar and 1550°C, which allows for high carbon conversion and cold gas efficiency. It is a slagging gasifier with membrane walls cooled with 54 bar evaporating water and insulated by the slag layer which minimizes heat losses and heat flux towards the membrane walls. A low sulfur South African coal (64.44% C, 3.95% H, 7.40% O, 1.49% N, 0.85% S, 9.20% H_2O, 12.67% Ash; 24.62 MJ/kg LHV) is used, pulverized and dried before feeding with a stream of warm air, heated by means of saturated water from the steam cycle HP drum. Oxygen is produced in a stand alone ASU, generating a 95% purity oxygen flow (stream 11). A pumped oxygen plant with a consumption of 325 kWh per metric ton of oxygen produced is used, according to data reported by Air Products [8]. Nitrogen produced in the ASU is partly used in lock hoppers (9) for coal feeding. Syngas produced is quenched to 900°C by means of cold (200°C) syngas recirculation (14). Molten slag entrained

by the gas stream solidifies and syngas is then cooled down in a convective syngas cooler, generating HP and IP steam. Most of the ash entrained in the raw syngas leaving the syngas cooler are removed in a high pressure, high temperature filter [9]. Syngas is then partly recirculated for quench, partly sent to a scrubber, where the remaining solids and soluble contaminants are removed. Syngas exits the scrubber at about 130°C (16) and after steam addition is sent to a high temperature water gas shift (WGS) reactor where CO is converted to CO_2 to obtain a H_2 to CO ratio of 3, optimal for methane production in the following methanation reactor. WGS reactor was calculated as a cooled reactor operating at 400°C (cheaper solutions based on uncooled reactors and syngas bypass can be adopted, with limited consequences on overall plant performance) and the flow rate of steam added was calibrated to obtain the target H_2/CO ratio. Shifted syngas is cooled down by producing HP steam for an efficient recovery of WGS heat of reaction and by preheating water and clean syngas.

Carbon dioxide and hydrogen sulfide (H_2S represents essentially all the sulfur species providing that the small amount of COS produced in the gasifier is hydrolyzed in WGS reactor), are removed in the acid gas removal (AGR) section by means of a chilled-Selexol based process. Two absorption columns are used for this task: one for CO_2 absorption, the other one, using part of the CO_2 pre-loaded solution exiting the first column as solvent, for H_2S absorption. Most of the absorbed carbon dioxide is released in flash chambers and compressed to liquefaction, while H_2S is desorbed in a stripper and sent to a Claus unit for elemental S production. Claus off-gas, rich of CO_2 and containing small amounts of unconverted sulfur species (95% of H_2S is assumed to be converted into S, considering the H_2S/CO_2 ratio at Claus inlet [10]) are firstly sent to a hydrogenation/hydrolysis catalytic reactor where the remaining sulfur is reduced to H_2S and then recycled back to the H_2S absorber inlet. Hydrogen sulfide is hence reabsorbed and CO_2 is not vented, increasing the overall carbon capture ratio of the plant.

Syngas exiting the Selexol unit has a hydrogen sulfide content of 10 ppmvd, while concentrations below 0.1 ppm are required to avoid fast poisoning of the following methanator Nickel-based catalyst. Therefore, an additional sulfur scavenging process is adopted after bulk removal in the Selexol unit. Sulfur scavenging processes are non-regenerative systems used to remove small quantities of sulfur compounds from gas streams. A number of sorbents (e.g. iron oxide, zinc oxide, nitrite solutions, polyamine solutions) can be used in such processes [11]. The most suitable one should be evaluated on the basis of economic analyses, highly dependent on the syngas composition. An already well known alternative, when very deep gas desulfurization is required, is the chilled methanol-based Rectisol process, which seems to be preferred in industrial practice. For example, in the Great Plains Synfuels Plant in North Dakota (USA), the only commercial synthetic natural gas (SNG) from coal plant, a Rectisol process is used for H_2S and CO_2 removal, producing a 20 ppb H_2S feed for the methanator [12]. However, Rectisol process is characterized by high complexity and energy consumptions and a system based on Selexol + sulfur scavenging seems preferable in plants like IGFCs, aiming at very high efficiencies.

Clean syngas from AGR unit is heated and sent to a methanation process (19), where carbon monoxide and hydrogen are converted into methane on a nickel-based catalyst according to the exothermic reaction (1):

$$CO + 3H_2 \rightarrow CH_4 + H_2O \qquad \Delta H^\circ_r = -206 \text{ kJ/mol} \qquad (1)$$

The main issues to be considered when designing a methanation process are [13,14]: (i) operating at conditions assuring fast kinetics and high CH_4 yields, (ii) avoiding catalyst poisoning due to the presence of sulfur species and chlorine, (iii) avoiding conditions, particularly critical at low temperatures and with low H_2/CO ratio, which can lead to catalyst deactivation for carbon deposition and nickel carbonyl formation, (iv) avoiding catalyst sintering at high temperatures. Temperature control is particularly important to respect these conditions and appears crucial considering the high exothermic character of methanation reaction. Processes proposed by manufacturers are generally based on fixed bed reactors operating between 250 and 700°C and using cooled product gas recycling (Lurgi process, TREMP of Haldor Topsøe and HICOM process of British Gas Corporation), cooled reactors (Linde process) or steam addition (RMP process and ICI process) to limit temperature increase along the reactor [14]. Steam addition also contributes increasing hydrogen and oxygen content in syngas, reducing the risk of carbon deposition. In all these processes, other lower temperature adiabatic reactors with intermediate cooling follow the first high temperature reactor to increase methane yield, which is favored at low temperatures for thermodynamic reasons. In the IGFC plant considered in this work, methanation is carried out in an adiabatic reactor with product gas recycle operating between 300 and 675°C, like in TREMP process proposed in [15] for applications in IGFCs. Product gas is recycled by means of a blower and is cooled by superheating high pressure steam. A single stage high temperature process is here adopted because high methane purities are not needed by SOFC and because using

lower temperature reactors would also require heat exchangers to heat the methane-rich gas up before entering the fuel cell.

The fuel cell is an Intermediate Temperature SOFC (IT-SOFC) working at 800°C. It operates in a hybrid layout with a gas turbine, where the fuel cell system oxidizes the fuel replacing the combustor. SOFC operates at elevated pressure, as imposed by the gasification island, with beneficial effects for its potential and for the overall plant efficiency [4]. The air stream feeding the cathode, exiting the compressor at about 590°C (2), is heated up to 700°C (a maximum ΔT of 100°C across the fuel cell was assumed), by recycling air exiting the FC cathode by means of ejectors. Ejectors allow preheating the airflow, eliminating or reducing the thermal duty of high temperature heat exchangers, following an approach already proposed by some manufacturers for future low-cost hybrid plants [16,17]. Thanks to the efficient cell cooling allowed by the reforming reactions taking place in the FC module, the amount of heat released to the air flow can be handled with the stipulated temperature rise (100°C) limiting the airflow (stream 3) to 437 kg/s (vs. 2033 kg/s found in previous configurations for the same power output [2,3]). This allows operating the FC with a higher air utilization factor (about 46%), with beneficial effect on the overall plant efficiency, as classically shown [7,18].

Anode exhaust gas (21) is cooled by producing high pressure superheated and reheated steam and sent to a post-SOFC high temperature WGS reactor, where carbon monoxide is converted into CO_2 and H_2 by reacting with water. Steam to CO ratio at anode outlet is higher than 8 and high CO conversions can be obtained in a single stage high temperature reactor without further steam addition. Gas exiting the post-anode WGS reactor is cooled down to nearly ambient temperature and sent to a post-SOFC CO_2 absorber using Selexol as physical solvent. The complete acid gas removal process is hence made of an H_2S absorption column and two (pre- and post-SOFC) CO_2 absorption columns. Hydrogen-rich gas released from the Selexol process is compressed, heated up (23) and burned with cathode exhausts increasing the gas turbine cycle temperature and hence its efficiency and specific work as described previously. Fuel utilization in the fuel cell was calibrated to obtain a hydrogen stream flow rate sufficient

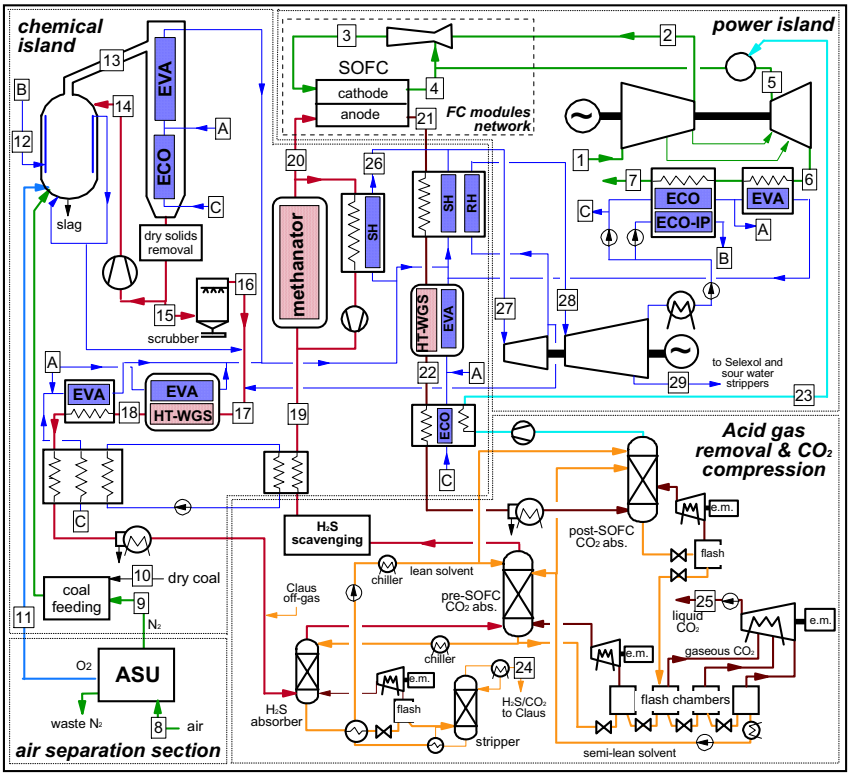

Figure 2: Layout of the proposed IGFC cycle using a methanation reactor and hydrogen firing in the combined cycle.

to reach a turbine inlet temperature typical of state-of-the-art gas turbines.

A single pressure level with reheat steam cycle (130/54 bar, 565/565°C) was selected for heat recovery. Most of the high temperature heat (over 350°C), which can be used for evaporation, superheating and reheating, is obtained by gasifier syngas cooling (121 MW), followed by methanation recycle (91 MW), anode exhaust cooling (50 MW) and GT flue gas (27 MW). In Fig.2, superheaters and reheater are positioned to recover heat from clean syngas, while gasifier syngas is cooled by evaporating steam in order to limit the heat exchangers surface exposed to high dust environments. Different configurations could be however adopted to increase plant operability and economics. A well designed heat recovery steam cycle, effectively exploiting high temperature heat, is anyway important for plant performance, considering the rather low cold gas efficiency of this layout as discussed further on.

Simulation of the presented power cycles has been carried out by a proprietary computer code (GS) developed by the Gecos group at the Department of Energy of Politecnico di Milano to assess the performance of gas/steam cycles and fuel cell systems [19]. The plant scheme is reproduced by assembling in a coherent network the different components selected in a library containing over 20 basic modules, whose models have been previously implemented. Built-in rules allow predicting turbomachines (gas and steam turbines, compressors) efficiency as a function of their operating conditions. Mass and energy balance of the Selexol and CO_2 compression processes were calculated with ASPEN Plus [20] using the PC-SAFT and the Peng-Robinson equations of state respectively. As to the thermodynamic model for Selexol propose in Aspen, the constant term (the one independent from temperature) of the binary interaction parameter equation were varied for each Selexol-gas couple to match the solubility data at ambient conditions reported in [21]. The resulting thermodynamic model was satisfactorily validated against proprietary data of a syngas treating process provided to Politecnico di Milano by UOP and the electric consumption of Selexol and CO_2 compression processes obtained in this work (416 kJ_e per kg of compressed CO_2) are consistent with values reported in [9] for a more common IGCC plant with CO_2 capture (430 $kJ_e/kgCO_2$).

4. Results

The outcomes of the simulation carried out for the described plant are presented are summarized in Tab.1 (third column) and compared with the other IGFC and IGCC plants assessed in [3,5]. The characteristics of the main streams are shown in Tab.2. The proposed power cycle achieves 51.6% net electrical efficiency (LHV base) with 95.3% carbon capture, a result which is about 4.5% points better than what was achieved with previous comparable configurations, not exploiting the modifications introduced here (methanation reactor and H_2 firing) [4]. The efficiency decay featured by the IGFC with CO_2 capture with respect to the cycle without capture is limited to 2.7% points, about one third of the value obtained in [4], demonstrating a more efficient thermodynamic integration among the energy conversion devices composing the power cycle.

Such a performance is obtained despite a rather low cold gas efficiency: SOFC fuel input in terms of LHV is in fact about 60% of the inlet coal LHV (against the 76.3-78.1% of the reference cases), the remaining 40% being converted into sensible heat. Most of the CGE reduction occurs in the methanation process where heat generated by the exothermic reaction is partly recovered by steam superheating and partly remains as sensible heat in the high temperature syngas. However, heat of methanation reaction is converted back into chemical energy in the SOFC by the endothermic steam reforming reaction, counterbalancing the CGE reduction.

The power balance shows that 52.5% of gross power is produced by SOFC, still a high value thinking to the expected high specific costs of fuel cells but 5-10% lower than what was found in [3]. The gas turbine generates a relatively small contribution to the power output, but, differently from other cases, its operating parameters (TIT and pressure ratio) are comparable with those of advanced aero-derivative commercial units, while air flow rate and power are respectively 80% and 30% larger than the largest simple-cycle aero-derivative machine now on the market [22]. Hence, a lower specific work also results, due to the extra power required by the compressor to drive the cathode ejector and to compress the oxygen used in the SOFC, which is not expanded in the turbine.

As anticipated, the SOFC works with a high air utilization fraction (46.5%). However, from the point of view of cell efficiency, the negative effect of a lower minimum oxygen fraction at cell outlet (8.1% vs. 18%), reducing the ideal Nernst voltage, is more than counterbalanced by the positive effects of operating at high pressure and with a limited fuel utilization and the resulting cell voltage is 0.812 V vs. 0.747 V of the reference IGFC with CO_2 capture.

As to the combustion process taking place downstream the SOFC, the lower oxygen fraction at cathode exhaust should be high enough for efficiently completing the H_2 combustion, leaving a O_2 fraction at combustor outlet of

about 2.3%. Another consequence is the low stoichiometric flame temperature (1910 K), which leads to low NO_x emissions obtained without fuel dilution or post-combustion selective catalytic reduction SCR.

Table 1 – Performance of the IGFC plant assessed in this and in previous works.

	IGFC			IGCC
Source	[5]	[3]	this work	[5]
CO_2 capture	no	yes	yes	no
Methanation and hydrogen firing	no	no	yes	-
SOFC pressure, bar	23.0	20.0	34.15	-
Single passage fuel utilization, %	26.0	70.0	74.0	
Overall fuel utilization, %	60.1	89.2	74.0	-
Air utilization, %	25.9	16.1	46.5	-
SOFC potential, V	0.808	0.747	0.812	-
GT compressor pressure ratio	25.5	22.1	38.7	17.0
TIT, °C	1335	659.4	1329	1335
TOT, °C	568.0	187.0	451.0	590.1
Flow at GT compressor inlet, kg/s	428.7	1026	264.7	546.0
Anode recirculation, % of SOFC inlet flow	75.9	78.3	-	-
Electric power, MW_e				
SOFC	183.8	331.4	305.9	-
Gas turbine	246.3	27.9	82.77	328.6
Steam turbine	136.6	161.5	193.9	207.2
Air separation unit (ASU)	-36.17	-45.55	-36.18	-36.17
Dilution N_2 compressor[a]/H_2 compressor[b]	-	-	-1.12[b]	-34.66[a]
Lock-hoppers N_2 compressor	-4.56	-	-4.56	-4.56
Syngas cooling recycle blower	-1.20	-1.24	-1.20	-1.20
Methanator recycle blower	-	-	-3.42	-
Steam cycle pumps	-3.06	-3.91	-3.64	-3.44
Acid gas removal and sulfur recovery	-0.35	-0.35	-13.45	-0.35
CO_2 compression	-	-15.22	-21.79	-
Auxiliaries for heat rejection	-1.90	-2.81	-3.16	-2.99
Pulverizers and coal handling	-1.79	-1.79	-1.79	-1.79
Slag handling	-0.49	-0.49	-0.49	-0.49
Miscellaneous BOP	-1.42	-1.42	-1.42	-1.42
Net power output, MW_e	515.7	447.8	490.4	448.7
Fuel input LHV, MW_{th}	950.0	950.0	950.0	950.0
CGE, %	78.07	76.27	59.98	78.07
Carbon capture ratio, %	0	97.50	95.33	0
Net LHV efficiency, %	54.29	47.14	51.62	47.24
Efficiency penalty, % points	-	7.15	2.67	-
Specific CO_2 emission, g/kWh	629.6	21.1	30.9	723.3

5. Conclusions

An advanced IGFC plant with methanation reactor and hydrogen firing was assessed in this work. Methanation and hydrogen combustion were considered to overcome the main limits of other IGFC configuration assessed by the authors in previous works, linked to the limited efficiency of the bottoming gas cycle. Even though high complexity and integration characterize the assessed layout, a considerable improvement in plant efficiency were obtained while significantly improving the expected gas cycle economics. For a 95% CO_2 capture ratio, a net plant efficiency of 51.6% was in fact calculated, 4.5% points higher than the reference IGFC.

Acknowledgments

John Bøgild Hansen of Haldor Topsøe is greatly acknowledged for the fruitful discussion on the application of the TREMP process in IGFC plants.

Table 2 – Temperature, pressure, flow rate and composition of selected points of the plant shown in Fig.2.

point	T,	p,	G,	M,	Molar composition, %								
	°C	bar	kg/s	kmole/s	CH_4	CO	CO_2	H_2	H_2O	Ar	N_2	O_2	H_2S
3	700	34.84	437.5	15.30			0.03		1.12	1.00	83.71	14.14	
4	800	34.15	405.3	14.30			0.03		1.20	1.07	89.61	8.09	
5	1419.5	33.13	193.0	7.05			1.20		10.65	1.34	84.56	2.25	
6	451	1.05	247.1	8.90			0.96		8.64	1.25	83.04	6.11	
18	400	40.99	100.4	4.18		13.93	28.60	42.77	7.19	0.65	6.65		0.21
19	300	35.55	39.02	3.20		20.68	4.42	64.09	0.06	0.93	9.82		
20	675	34.84	39.03	2.10	26.29	3.75	8.26	20.43	24.87	1.42	14.98		
21	800	34.15	71.23	3.20		5.94	19.17	16.11	48.05	0.93	9.82		
22	400	32.44	71.23	3.20		0.97	24.13	21.07	43.08	0.93	9.82		
23	250	40.98	14.10	1.09		2.80	4.77	61.97	0.06	2.51	27.89		

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